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# Diagnostics of Low Temperature Plasmas by Optical Means

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In this paper a short overview of the diagnostics of low temperature plasmas by optical means is presented. It is described how the results of the optical measurements, namely the intensities and profiles of the spectral lines can be used for measuring the low temperature plasma parameters - temperatures, electron density, plasma velocity, magnetic and electric field in the plasma etc. Discussed are the strengths and limitations of these methods.

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## 1 Introduction

Plasma diagnostics can be performed using very different methods: by application of Langmuir probes, mass spectroscopy, X-ray spectroscopy (for high temperature plasmas), analysis of electrical properties of the plasma. Most widely however, the optical methods are used, namely study of the optical emission of the plasma or its absorption of external light. There are also more sophisticated methods of plasma diagnostics using laser light scattering or interferometry, but this article will take into account from many possible laser methods only the Laser Induced Fluorescence (LIF), where the analysis of the signal is done in nearly exactly the same way as the analysis of results from emission or absorption spectroscopy.

There are two possible approaches to determining the plasma parameters from optical signals - one using the total signal of the atomic/ionic line or molecular band (total emitted or absorbed light intensity), or by analysis of the line profile. The results of those analyses can provide most of the plasma parameters, depending on the state of the plasma. It is easier if Local Thermal Equilibrium can be assumed because then several of the plasma parameters can be calculated using the equilibrium equations.

In this paper only a short descriptions of the methods and the citations from other publications are provided, due to the article length constraints.

## 2 Plasma parameters which can be determined by optical means

Some of the plasma parameters can be obtained by straightforward analysis of optical signals, some other can be calculated assuming some equilibrium conditions (either thermal equilibrium or more complicated ones, like collisional-radiative model). The straightforward methods include:

- densities (absolute or relative) of given emitters in particular state of excitation
- excitation temperatures for a given subsystem of the studied plasma
- velocity distribution functions for different species in the plasma, which in the case of Maxwellian distribution can also be described using the parameter called temperature
- bulk velocity of plasma or its subspecies (e.g. atoms or ions, which do not always exhibit the same velocity)
- electron density

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- values of external and internal electric and magnetic fields.

From above mentioned parameters some additional ones can be derived, like total density of the plasma, plasma pressure etc.

### 3 Methods of optical measurements

In this article only emission, absorption and LIF spectroscopy are discussed. The emission spectroscopy is the easiest and quickest to perform and it is also a non-intrusive method. Analysis of emission signals is performed using spectrometers of given spectral resolution. Signals measured with high resolution spectrometers can be used for profile analysis, measured with lower resolution are suitable for total line intensity measurements.

The greatest problem of the emission spectroscopy method is that it does not allow for performing local measurements, and the light which arrives to the spectrometer is integrated over the line of sight. If the plasma parameters can be considered uniform over the line of sight the interpretation of spectra does not differ from local measurements, but if there are any gradients along this line the signal will not give directly the required information. There are many works where such integrated signals are used to determine the "mean" or "effective" values of studied parameters (which can be far from reality in many cases). Local values of emission coefficients can be obtained from the integrated signals if certain symmetries can be assumed but the results have additional uncertainties.

Emission spectroscopy is also restricted to species in the plasma which actually emit radiation, so by emission spectroscopy one cannot measure for example the density of atoms in the ground level. Absorption spectroscopy has not this limitation, but is also a non-local measurement. Absorption needs also an external light source (preferably wavelength adjusted), and is very limited in case of luminous plasmas, because the plasma emission can be strong enough to make the absorption in the medium invisible. Absorption spectroscopy is also an intrusive method and can influence the properties of the plasma.

Laser Induced Fluorescence is a local measurement, where the information obtained originates from the cross-section of the laser beam and the optical axis of the fluorescence observation. LIF can give information about both ground and excited states and is very sensitive because of the heterodyne detection which can be used in such cases. On the other hand, it is also much more complicated from experimental point of view, and is also nearly as intrusive as the absorption spectroscopy (less, because due to its sensitivity much less of the light energy has to be absorbed to yield the observable signal). In principle it does not need any spectrometer, but in most cases at least a filter is needed to distinguish the LIF signal from the plasma emission.

Both emission and LIF spectroscopy are much more complicated to analyze in case when the medium is strongly absorbing. LIF in such case loses also its locality, because one has to take into account not only the interaction of the laser light with plasma in the observed volume, but the analysis should also include absorption of the laser light on its way to the observed volume, and the absorption of the LIF signal along the line of sight. In the case of emission spectroscopy, where tomography methods are required they can be upgraded to take absorption into account as well, but the analysis is very difficult and the results are often uncertain.

## 4 Analysis of the total line intensities

### 4.1 Description

The total emission coefficient of the transition from the level 2 to level 1 can be described using the following equation:

$$\epsilon_{tot} = A_{21}N_2h\nu_{21} \quad (1)$$

where  $N_2$  is the density of the emitters in the excited level (2),  $A_{21}$  is the respective Einstein coefficient of the transition and  $\nu_{21}$  is the frequency of the transition. The plasma parameter of interest can be directly the density of the emitters  $N_2$ , which can be obtained from measurements of properly selected lines if the absolute emission coefficient is known (radiance calibration of the instrument required). In most cases this density is used to determine the excitation temperature, described by the Boltzmann equilibrium:

$$N_i = N_{tot} \frac{g_i \exp(-E_i/kT)}{\sum g_k \exp(-E_k/kT)} \quad (2)$$

where  $g_i$  is the statistical weight of the level  $i$ ,  $E_i$  is the excitation energy of the level and  $T$  is the excitation temperature of the given subsystem. In principle this formula can be used to obtain the temperature from absolute emission coefficient of one emission line.

Using the absolute intensity of the emission lines requires very precise calibration of the measurement and the knowledge of partition function (denominator in the eq. 2). The uncertainty of such a measurement is nevertheless pretty high, especially because the partition function depends on the plasma parameters, so much more precise is to calculate temperature using several lines from the same subsystem (e.g. atomic lines of the same atom, molecular lines from the same vibrational-rotational band) applying the Boltzmann diagram, where the logarithm of  $I_{tot}/g_i A_{21} \nu_{21}$  is plotted against the excited level energy  $E_i$ . This way one can avoid both the partition function and absolute calibration of the line intensity and use only the relative spectral response calibration.

To improve the measurement many considerations must be taken into account. In principle two lines can suffice, but for more precise determination of the uncertainty of the temperature determination at least four should be used, with significantly different excitation energies (at least equal to the  $kT$  - temperature expressed in units of energy). The lines should be isolated, or at least easily resolved and it's advantageous if they are close in wavelength, because in such cases the spectral response of the equipment differs only slightly, so the uncertainty of intensity calibration is small.

The Boltzmann plot can be used only in the case if the Boltzmann equilibrium between the excited states of the subsystem can be assumed. In most cases the ground level is not in thermal equilibrium with excited states (in wall-stabilized arcs working in argon or helium), so the determination of temperature or total density of the species can be difficult. It can still be done, but mostly with the help of collisional-radiative models. The temperature derived from atomic or ionic lines is the so-called excitation temperature, which in partial LTE plasmas is close to the electron temperature.

#### 4.2 Examples of the Boltzmann plot technique

Boltzmann plot can be used both for atomic/ionic lines as well as for molecular ones, if the rotational molecular structure can be resolved in the experiment. In references [1, 2] two sets of lines (O I and N I) suitable for temperature determination are proposed. The nitrogen and oxygen lines are very useful for plasma diagnostics because even in plasmas where those elements are not introduced, traces of them (as of hydrogen as well) can still be found and used for plasma diagnostics.

The same can be said about the nitrogen molecular bands, both of the neutral molecule itself, and of the molecular ion. The measurement of temperature based on the molecular ion spectrum can provide data for lower temperatures than those obtained from the measurement of atomic lines. In the case of molecule, though, the temperature is obtained mostly from rotational subsystem, which provides rotational temperatures, commonly assumed to be close to the gas temperatures. The Boltzmann plot for such temperature measurement in the fringes of the cascade arc is shown in reference [3]. In this publication Boltzmann plots are shown corresponding to two different distances from the arc axis.

#### 4.3 Example of an analysis of unresolved molecular bands

In the case of molecules, the rotational structure cannot always be resolved. One can also be interested not only in rotational temperature but in vibrational one, which is much more difficult to measure, because the vibrational molecular transition consists of a full band of lines (not one). In that cases the molecular band is not resolved but modeled and compared/fitted to the experimental profile. The situation in this case is much more complicated, because it is not easy to find a molecular band completely undisturbed by other bands, but it can be done and the example is presented in reference [4] (similar spectra were calculated also earlier, for example in [5]).

### 5 Analysis of the line profiles

Analysis of line profiles requires the detection of the profiles with high resolution, so the width of the line must be much larger than any limits arising from the equipment (instrumental profile for emission, laser linewidth for LIF and absorption). The profile of the atomic/ionic line is determined by following components:

- internal structure of the line: fine/hyperfine structure, isotope shift (if there is a mixture of isotopes), splitting and shift of the line due to external electric or magnetic fields
- broadening originating from: natural linewidth, pressure broadening, power broadening (in the case of laser spectroscopy) - all Lorentz and quasi-Lorentz components,
- Maxwellian distribution of emitters velocity - Gaussian profile, in the case of non-Maxwellian distribution the profile is non-Gaussian or at least many-Gaussian
- line shifts due to collisions or Doppler shift because of nonzero bulk velocity of the plasma
- instrumental profiles, which can be of many shapes: Lorentz, Voigt, triangle, Airy (in the case of Fabry-Pérot spectrometers).

Contributions originating from above described components to the observed profiles may significantly differ from each other, even by orders of magnitude. Unfortunately, sometimes the most interesting parameter isn't the largest one, and then resolving the contributions can be difficult and the plasma parameter derived from the profile can have large uncertainty.

In the case of very simple profiles with one or two predominant factors the line profile can be fitted using Gaussian/Lorentz/Voigt function (one or more) and the plasma parameters can be calculated from the fit results. In the case of very complicated profiles, especially when the profile is measured by emission spectroscopy and the symmetry of the medium cannot be assumed or the medium is not optically thin, the profile can be still modeled from assumed plasma properties and used to verify the plasma model. In this case fits are rarely usable due to too many free parameters of the fit, often having similar influence of the profile shape.

### 5.1 Example of simple Doppler broadening and shift

This example, is a simple profile of an argon 810.37 nm line, where the predominant broadening factor is the Doppler broadening of the line, and the profile is nearly exactly Gaussian, so the Maxwell velocity distribution of the atoms can be assumed (ref. [6]). The line is shifted against the "static" wavelength because of the nonzero bulk plasma velocity. The profile is measured using LIF in optically thin medium, so the measurement is local and undisturbed by the rest of the plasma. In this case the Doppler temperature and the plasma velocity can be easily determined from the width and shift of the line:

$$T \propto \Delta\nu_{\text{HWHM}}^2, v \propto \Delta\nu_{\text{shift}} \quad (3)$$

The plasma studied in this example is a hypersonic argon/nitrogen jet. The velocity distribution of the atoms is Maxwellian, but the parameters of the distribution depend on the direction in space (they are different along the plasma axis and the direction perpendicular to the plasma axis).

### 5.2 Example of the many-Doppler profiles

In the case of hypersonic jets the Maxwell velocity distribution can be assumed, but the gas can consist of two or more components with different temperatures and velocities, so the profile can consist of several Gaussians. An example of such situation is shown in reference [7], where there are two velocity components in a supersonic argon jet. In this case the respective temperatures and velocities of the components can be calculated in the same way as in the previous example. The profiles are also measured by the LIF technique, using an argon line.

### 5.3 Example of the complicated, reabsorbed profile from nonsymmetric medium

These profiles, shown in reference [8] are the emission profiles of the argon 763 nm line measured from pure argon hypersonic jet, the same as described in [6]. In this case the observed line was strongly reabsorbed and the spectra were recorded with the help of a very precise Fabry-Pérot spectrometer in the direction perpendicular to the jet axis. The medium couldn't be considered symmetric because of the jet expansion - the jet was expanding symmetrically, but the Doppler shift of this expansion, being a scalar product of the velocity vector of the expansion and the direction of the optical axis, was not symmetric.

In such complicated case (nonsymmetric medium and reabsorption) the profile depends on the whole spatial distribution along the line of sight of several plasma parameters - density of emitters, density of absorbers, velocity of the plasma, Doppler temperature of the emitters and of absorbers (those two were assumed equal). The approach was to reproduce the observed spectrum from the model and verify, if the model agrees with the laser measurements performed in [6]. Similar methods are used to verify the models of the plasma by the help of the emission spectra - reference [9].

#### 5.4 Example of the measurement of the external electric field in the plasma

In this example (reference [10]) the spectra were recorded using the optical emission spectroscopy and were generated in a barrier discharge plasma working in the mixture of argon and hydrogen. The measured line was the hydrogen Balmer alpha line (656.2 nm), exhibiting the linear Stark effect. In this case, the line was splitted into several  $\pi$  and  $\sigma$  polarized components by the external electric field. The profiles were fitted to the experimental spectra of separated  $\pi$  and  $\sigma$  components using the following assumptions: each component consists of two Gaussians, one generated by fast, the second by slow hydrogen atoms, both centered at the same wavelength; the location of the components is determined by electric field and the relative intensities of the components are taken from the theory of Stark splitting. This means five free parameters for each pair of spectra ( $\pi$  and  $\sigma$  components were fitted simultaneously). The measurement gives the values of electric field strengths corresponding to different layers of the plasma source.

#### 5.5 Example of the influence of the electron density and temperature on the line profile

The charged particles in the plasma influence the line profiles of emission lines. The magnitude of it depends on the emitted line, but one of the strongest influences is visible for the hydrogen lines. This is also a Stark splitting as in the previous example, but from time-dependent microfields in the plasma, divided in two domains - fast changing microfields of electrons and slowly changing microfields of ions. This influence produces the so-called Griem profiles, which can in some cases be approximated by Lorentz profiles.

Width and shift of atomic lines in the plasma can be correlated with the density and temperature of electrons, which can lead to the derivation of such plasma parameters from the width of the emitted line. Commonly used as the probe of the electron density are the hydrogen lines, because of their large widths, measurement of which does not need the very high resolution.

To use the experimental linewidth for obtaining the electron density one has to know the dependence from either theoretical calculations or experimental calibration. Figures in reference [11] show the theoretical profile of the hydrogen Balmer alpha line for different electron densities (the Doppler broadening is in this case neglected). It is visible, that for electron densities higher than the threshold value of  $10^{14} \text{ cm}^{-3}$  (for low values of the Doppler broadening)  $H_{\alpha}$  width is a good measure of electron density.  $H_{\alpha}$  is a very useful probe for electron density due to its large width and quasi-Lorentzian shape, but for lower electron densities much preferred is even wider  $H_{\beta}$ . This line is a bit more difficult to analyze experimentally due to its two-maximum shape, but on the other hand this profile can be helpful for determining the homogeneity of the observed plasma.

#### 5.6 Example of the estimation of the magnetic field in the plasma

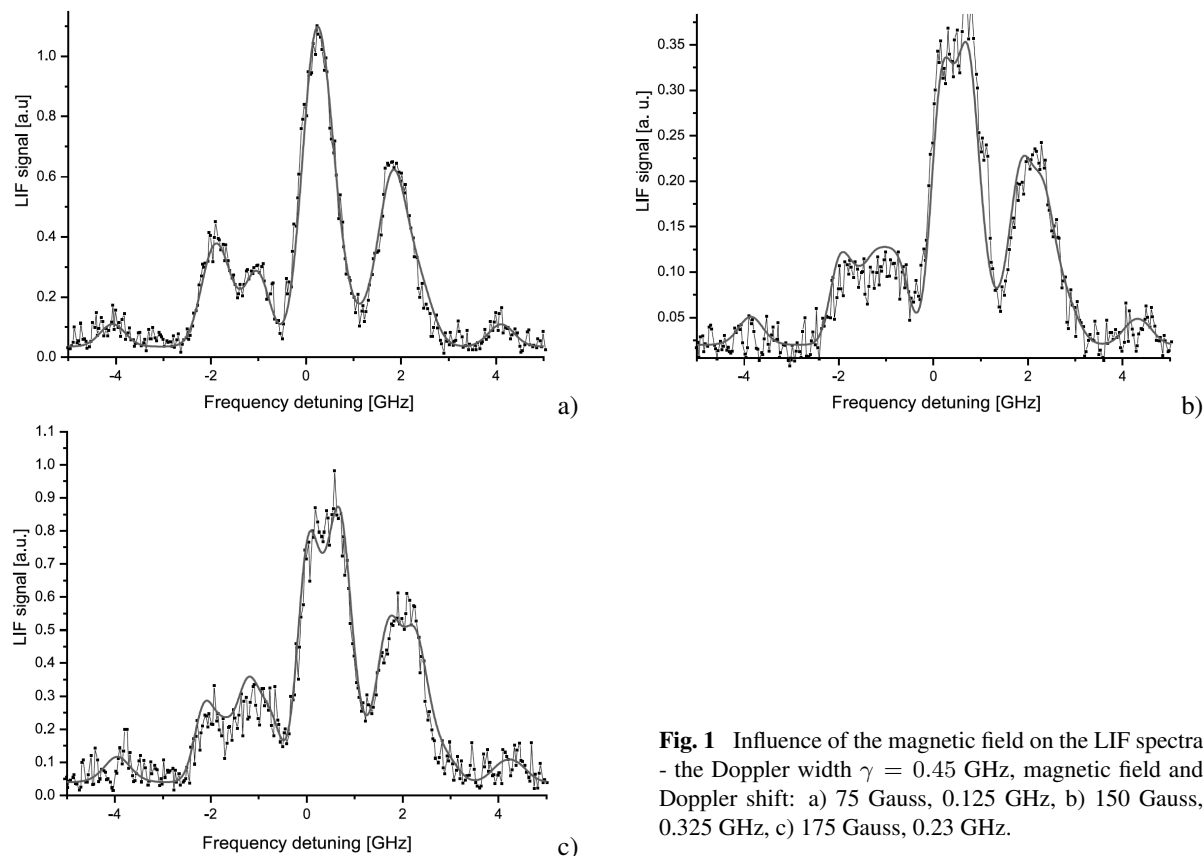
The spectra presented in figure ?? show the influence of the magnetic field on the LIF spectra of the Xe I 823.163 nm line. This line is very complicated due to the fact, that xenon is a mixture of both even and odd isotopes. The lines originating from odd isotopes (with 1/2 and 3/2 nuclear spin) exhibit visible hyperfine structure and all of the isotopes have non-negligible isotope shift.

The mechanisms that have to be included in the analysis of those spectra in addition to hyperfine structure and isotope shifts of all the xenon isotopes include:

- Doppler broadening of each of the components, which can, but need not, result in the Gaussian shapes of the components (the velocity distribution functions can be non-Maxwellian) and overall Doppler shift of the spectrum due to the plasma bulk velocity
- saturation from the applied laser power - the broadening is negligible with comparison to the Doppler broadening, but the saturation affects the relative intensities of the hyperfine components

- splitting of the components due to the magnetic field in the plasma.

The splitting of the lines cannot be observed directly because the magnetic field is too weak to shift the lines significantly in comparison to the linewidth, but it can and does change the profile, for example the width of the main peak or the relative intensities of the components. This dependence is somewhat complex, but still can be tabulated and used for the estimation of the magnetic field in the plasma.



**Fig. 1** Influence of the magnetic field on the LIF spectra - the Doppler width  $\gamma = 0.45$  GHz, magnetic field and Doppler shift: a) 75 Gauss, 0.125 GHz, b) 150 Gauss, 0.325 GHz, c) 175 Gauss, 0.23 GHz.

## 6 Conclusions

As presented in the examples described above, most of the plasma parameters can be derived from the analysis of optical measurements. Even if the profile of the emission/absorption/LIF line is too complicated to derive the plasma parameters directly, one still can use the experimental data to verify the plasma models and compare with other kinds of measurements.

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